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The disintegration of In^{116}

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With 5 figures in the text

1. Introduction

In 1934 AMALDI, D'AGOSTINO and SEGRE (1, 2) discovered that slow neutrons induced activities with half-lives of 13 seconds and 54 minutes in indium. By deflecting the outgoing radiation in a magnetic field they were able to show that negatrons are radiated by the 54 min. activity. About the same time SZILARD and CHALMERS (3) reported that they had observed two activities of half-lives 1 hour and 3.5 hours in indium irradiated by neutrons. They pointed out that as indium only has two stable isotopes viz. In^{113} and In^{115}, one isotope must be activated with more than one period.

Since then the 13 sec. and 54 min. activities of indium have been investigated extensively, by different methods, often with conflicting results. It was shown (4) that the 54 min. activity was also induced by the fast neutrons from lithium bombarded by 6.3 MeV deuterons. LAWSON and CORK (5) showed that both activities are induced in In^{115} and belong to isomeric states of In^{116}. The activation cross section of In^{115} for thermal neutrons was determined by HOUTERMANS (6) and by SEREN, FRIEDLANDER and TURKEL (7). The last named authors found 52 barns for the 13 sec. activity and 145 barns for that of 54 minutes.

The radiations from In^{116} were investigated by means of a Wilson chamber by GAERTTNER, TURIN and CRANE (8) who found upper limits of respectively 1.3 MeV and 3.2 MeV for the energy of the negatrons emitted by the 54 min. and 13 sec. activities. They also found indications of a γ-radiation of 1 MeV, and possibly a weaker γ-radiation of higher energy. Using the same method BROWN and MITCHELL (9) determined the upper limit for the continuous electron distribution of the 54 min. activity as 1.45 MeV. Further Wilson chamber work showed (10) that no γ-radiation accompanies the 13 sec. activity, and by using a large hydrogen filled Wilson chamber CURTIS and RICHARDSON (11) indicated the existence of γ-lines at 0.2, 0.4, 1.0, 1.3, and 1.8 MeV, with relative intensities of 0.1, 0.3, 1.0, 1.0, and 0.3 resp., in the 54 min. decay. The γ-rays at 1.0, 1.3, and 1.8 MeV were confirmed by CORK and LAWSON (12), who also determined the upper limit of the continuous β-spectrum as 0.85 MeV. The Wilson chamber measurements could not determine whether the β-spectrum was simple or complex (8, 9).

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In a later investigation CURTIS and RICHARDSON (13) also found 0.85 MeV for the upper limit and determined the energies of the γ-rays as 0.17, 0.358, 0.57, 1.02, 1.40, and 1.85 MeV.

In^{116} has also been investigated by means of the absorption and coincidence techniques. The first measurements of MITCHELL and LANGER (14), in which they applied the absorption technique and measured coincidences between the Compton electrons from an aluminum radiator, seemed to indicate that the 54 min. activity was accompanied only by a monochromatic 1.39 MeV γ-radiation. Later experiments by LANGER, MITCHELL and MCDANIEL (15) showed a γγ-coincidence effect, however, and indicated that each disintegration was accompanied by about two gamma quanta. Although the βγ-coincidence measurements seemed to indicate a simple β-spectrum, the β-absorption curve showed a break at 0.8 MeV.

LAWSON and CORK (16) did absorption experiments on 13 sec. In^{116} and determined the upper limit of the continuous β-spectrum as 3.2 MeV. Their results were later evaluated carefully by BLEULER and ZÜNTI (17) and the upper limit determined as 2.95 ± 0.10 MeV. Absorption of the continuous β-spectrum of the 54 min. activity in copper (18) gave 1.0 MeV for the upper limit. MANDEVILLE and SCHERB (19) determined this upper limit at 0.7 MeV by βγ-coincidence and absorption measurements, and stated that the β-spectrum was simple.

A spectrometrical investigation by DEUTSCH, ROBERTS and ELLIOT (20) gave the γ-ray energies as 0.428, 1.12, 1.31, and 2.32 MeV (± 3 %), and by investigating the photo disintegration of Be by the In^{116} γ-radiation, HUGHES and EGGLER (21) found two groups of photo neutrons, which seemed to indicate that In^{116} emitted γ-rays with energies of 2.08 ± 0.04 MeV and about 1.8 MeV.

To account for the experimental data, disintegration schemes were proposed by MITCHELL and LANGER (10) and by CURTIS and RICHARDSON (13). The results previously obtained are summarized for convenience in Table 1.

As improved techniques for investigating the radiations of radioactive substances are now available, it was considered worth while to reexamine the disintegration of this isotope.

2. Source Preparation and Irradiation

Because of the large cross section of In^{115} for slow neutrons, and its relatively short half-life, strong samples of In^{116} can be prepared by irradiating chemically pure indium with slow neutrons outside the vacuum-chamber of a cyclotron. For this investigation, different sources were prepared for different purposes. For obtaining the continuous β-spectrum with the accompanying internal conversion lines, thin uniform sources were prepared by evaporating indium onto a backing of aluminum foil, 0.15 mg/cm^2 thick. In this way a number of sources were prepared, with thicknesses of 1.3, 0.7, and 0.22 mg/cm^2. Before irradiation these sources were ready mounted on bakelite rings, so that they could after irradiation be slipped into the spectrometer without delay.

For obtaining the photo- and Compton spectra, sources could be made very conveniently by casting slugs to fit exactly into the copper radiators to be used in the various spectrometers.

In a preliminary experiment indium was irradiated for about one hour, and

Table 1

The radiations of In^{116} as determined by various techniques.

Activity	β-spectrum	γ-radiation	Method	Ref.
54 min.	1.3 MeV simple		W	8
	1.45 MeV		W	9
		1.39 MeV	CA	14
	1.4 ± 0.1 MeV simple	0.2 0.4 1.0 1.3 1.8 MeV	W	10
		(0.1) (0.3) (1.0) (1.0) (0.3)	E β by S	
	0.85 MeV	1.0 1.3 1.8 MeV	E γ by W	12
	1.0 MeV		A	18
	0.7 MeV simple		CA	19
		1.8 2.08 ± 0.04	P	21
	0.85 MeV simple	0.17, 0.358, 0.57, 1.02, 1.40, 1.85	W	13
		0.428 1.12, 1.31, 2.32 ± 3 %	S	20
13 sec.	3.2 MeV			8
	3.1 ± 0.1 MeV	No gamma		10
	3.2 MeV			16
	2.95 ± 0.10 MeV			16+17

The figures in brackets are proportional to the intensities of the components. W = Wilson Chamber, A = absorption, C = coincidence method, S = spectrometer, P = photo disintegration.

a decay curve taken. The activity showed a simple logarithmic decay with a half-life of about 54 minutes. It was possible to follow this decay for almost six half-lives without it showing any deviation from a linear logarithmic plot, thus showing that no other activities are induced to any measurable extent. It was thus not necessary to do any chemical separation on the irradiated samples, and they could be used again and again. Even after a few months it was not possible to detect any measurable long-lived activity in them.

It was also possible to observe the 13 seconds activity, by limiting the irradiation to 1 minute, and measuring the half-live immediately afterwards. This activity could be followed for 4 half-lives.

No trace of the 4.5 hour activity first reported by SZILARD and CHALMERS (3) as a 3.5 hour activity, and which is actually due to an isomeric state (22) of In^{115} induced by fast neutrons, was observed.

3. The beta-spectrum

The high-transmission intermediate image spectrometer (23, 24) was used for investigating the β-spectrum. The Geiger-Müller tube used had a nylon window with a thickness of about 0.1 mg/cm^2.

It was found that after 2 hours irradiation an In-sample of 0.22 mg/cm^2 gave a counting rate of about 4 000 counts/min. at the highest point of the continuous electron distribution. It was therefore quite suitable for determining the form of the β-spectrum. After one activation it was possible to cover the continuous spectrum five times before the source became too weak. Each point was corrected for decay, and the five curves obtained could then be combined with due regard to their different statistical weights.

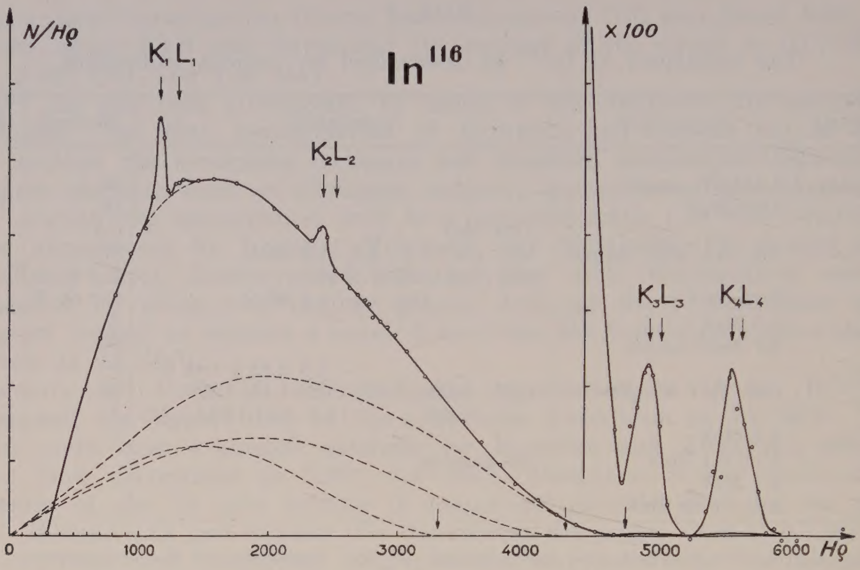

Fig. 1. The β-spectrum of In^{116}.

A Fermi analysis of the final curve showed that it was complex with at least three and possibly four components.

In order to determine the upper energy limits of the two components of highest energy as accurately as possible, the more energetic part of the continuous spectrum was measured by using source of 1.3 mg/cm^2 thickness. This allowed this part of the spectrum to be measured with very good statistical accuracy and the upper energy limits of the more energetic components to be determined more accurately. This data could then be used as an aid in analyzing the results obtained with the thinner source, and the component(s) of lower energy could consequently be better determined.

Altogether five completely independent runs were made, and the necessary Fermi analysis done on the results.

Apart from these investigations, the β-spectrum was carefully examined in detail for possible internal conversion lines by means of several runs.

By combining a number of runs obtained with the same source, Fig. 1 was obtained. It shows the K- and L-conversion lines of a γ-ray of energy 0.137 MeV ± 1 %, as well as the K-conversion lines of γ-rays at 0.409 MeV, 1.080 MeV and 1.208 MeV. The arrows marked K_2, L_2, K_3, L_3, K_4 and L_4 indicate the positions where one would, according to energies determined from the photo-spectrum, expect to find K- and L-conversion lines. It is clear that with the resolution used, it was not possible to resolve the K- and L-lines.

The broken curves are continuous distributions corresponding to three straight lines obtained in the Fermi-plot. Together they account for 94 % of the continuous spectrum, and have relative intensities of 51 %, 28 % and 21 %, beginning with the more energetic component. At energies smaller than 250 KeV a 6 % surplus of electrons deviates above the last straight line of the

Fermi diagram. As no γ-ray could be found to take care of a soft component of this energy, it seems probable that this surplus does not represent a fourth component, but is a result of straggling, back scattering etc. Such deviations have been observed with thinner source than that used here (25, 26).

The upper energy limits of the β-components, as determined from the Fermi analysis are 1.00 MeV (51 %), 0.87 MeV (28 %) and 0.60 MeV (21 %). The corresponding $H\rho$-values are indicated by arrows in Fig. 1.

4. The gamma-spectrum

Apart from the conversion lines found, the γ-spectrum of In^{116} was also examined by studying the Compton and photo-spectra. The copper radiators into which the sources were placed had a wall thickness of at least 1 mm, so as to exclude all possible β-radiation up to an energy of 1.9 MeV.

The Compton spectrum was taken with a lens spectrometer (27) and is reproduced by the continuous curve of Fig. 2. Special attention was paid to that part of the curve which lies between 5 000 and 8 000 gauss-cm, in an effort to bring to light the γ-ray at about 1.8 MeV, which has been reported by several authors (11, 12, 13, 21). It has been impossible to find any Compton edge corresponding to a γ-ray of this energy. It was also impossible to find any photoline corresponding to this γ-ray, and we have to conclude that if this γ-line exists, it follows less that 3 % of the disintegrations of In^{116}.

The Compton distribution shown in Fig. 2, is actually a combination of curves obtained during two runs, using a new source each time. These curves agreed very well with each other.

The arrows marked C_1, C_2 . . . C_6 indicated the expected positions of the Compton edges of the various γ-rays, which showed up as photo-lines in the photo-spectrum. The agreement between the two methods is very good. The Compton spectrum shows clearly that no γ-rays above the energy corresponding to C_6, i.e. 2.09 MeV are to be expected.

To study the photo-spectrum, a lead foil of 34 mg/cm^2 was placed on the Compton radiator and the whole energy region from the counter window cutoff ($H\rho = 300$ gauss-cm) to 2.5 MeV investigated with the lens spectrometer (27) (resolution 4 %) in order to search for photo-lines. After these had been localized they were taken up one by one in detail on the intermediate image spectrometer (23, 24), which was adjusted to give a resolution of 3.2 %. As this enabled a fairly accurate determination of their energies, it was then easier to get a more accurate determination by investigating just the lines themselves in the double focusing spectrometer (28). In order to enable intensity comparisons to be made later, the lines were taken up in pairs, the same line being measured each time together with another line. Owing to the comparatively rapid decay of In^{116} it was impossible to measure more than two lines with the same source.

The photo-lines corresponding to the γ-ray of 0.137 MeV of which the internal conversion line had been found in the β-spectrum, could not be measured in the double focusing spectrometer because the counter window absorption was too great. It was possible however to measure the photo-lines, due to this γ-ray, in the lens spectrometer.

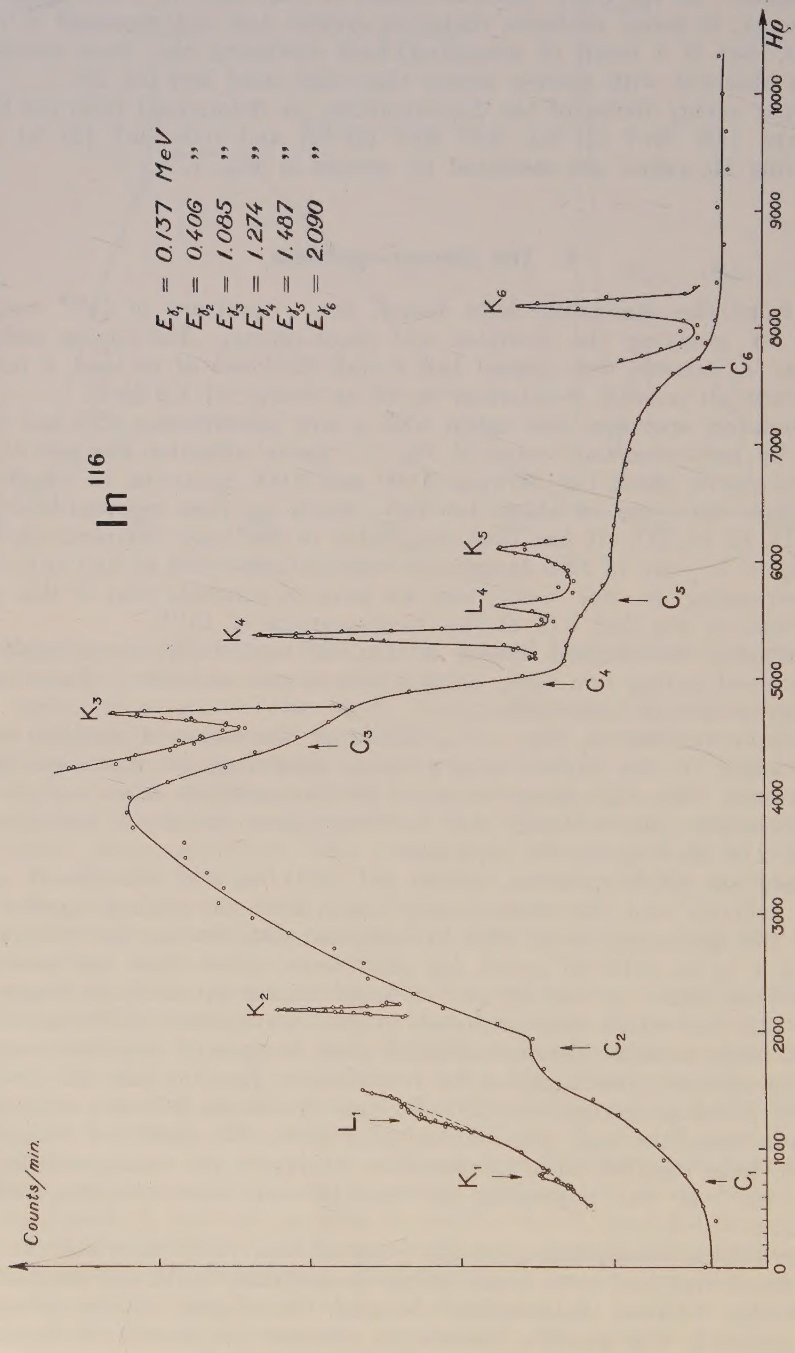

Fig. 2. The Compton and photo-spectra of In^{116}.

The various photo-lines are shown above the Compton spectrum in Fig. 2. The arrows marked K$_1$ and L$_1$ show the positions where one would expect to find the K and L photo-lines of a gamma ray of 0.137 MeV. As the figure shows the lines found with the lens spectrometer agree very well this figure, but the internal conversion line was obviously more suitable for an energy determination.

The other lines shown in Fig. 1 were taken on the double focusing spectrometer, and correspond to γ-rays with energies of resp. 0.406, 1.085, 1.274, 1.487 and 2.090 MeV. The accuracy of thesse energies is ± 0.5 %. The lines K$_1$ and L$_1$ were taken with a 34 mg/cm^2 lead radiator, K$_2$ with one of 8.7 mg/cm^2 and the rest with one of 20 mg/cm^2. The lines shown in Fig. 2 were taken with different sources of In^{116}, and their areas do not have any simple relation to their intensities.

The various energy determinations done on the γ-lines of In^{116} are summarized in Table 2, where S is the spectrometer described in 27, SS the intermediate image spectrometer 23, 24 and D stands for the double focusing spectrometer 28.

Table 2

The γ-rays of In^{116}.

Method	Spectrometer	Energy of γ-ray in MeV	Final energy in MeV
L photo-line	S	0.141 ± 0.006	
K » »	S	0.138 ± 0.004	
K internal conversion	SS	0.137 ± 0.001	**0.137 ± 0.001**
K conversion	SS	0.409 ± 0.006	
L photo-line	SS	0.400 ± 0.010	
K » »	SS	0.407 ± 0.010	
K » »	D	0.404 ± 0.003	
K » »	D	0.408 ± 0.002	**0.406 ± 0.002**
K internal conversion	SS	1.080 ± 0.01	
K photo-line	SS	1.082 ± 0.01	
K » »	D	1.084 ± 0.006	
K » »	D	1.085 ± 0.006	
K » »	D	1.085 ± 0.006	**0.085 ± 0.005**
K photo-line	S	1.280 ± 0.02	
K internal conversion	SS	1.268 ± 0.01	
K photo-line	SS	1.274 ± 0.01	
L » »	D	1.274 ± 0.008	
K » »	D	1.274 ± 0.006	**1.274 ± 0.006**
K photo-line	SS	1.461 ± 0.03	
K » »	S	1.484 ± 0.02	
K » »	D	1.487 ± 0.007	**1.487 ± 0.007**
K photo-line	S	2.097 ± 0.03	
K » »	SS	2.079 ± 0.02	
K » »	D	2.090 ± 0.01	**2.090 ± 0.010**

The energies of the various lines have been determined with respect to the ThC″ γ-line for which a Hρ-value of 10.000 ± 14 gauss-cm has been accepted (29).

5. The disintegration scheme of In^{116}

With the data, given above, available, it was possible to draw up a disintegration scheme for In^{116} which agreed with all the experimental facts. This scheme is shown in Fig. 3. According to it the 2.496 MeV level of Sn^{116} can be deexcited by the competitive emission of γ-quanta of 0.137 MeV and 0.406 MeV (or 2.090 MeV).

To determine what proportion of the 28 % disintegrations, feeding this level, goes via each of two alternative ways, the following procedure was followed: the ft-values for the three components were calculated, giving the results shown in Table 3. For this purpose use was made of theoretical curves kindly supplied prior to publication by Dr. ROSE (31). According to the ft-values given in Table 3 it seems that all the β-transitions belong to the symmetry forbidden class (32) with no change in parity. The 0.137 transition has therefore to occur between levels of the same parity, and according to the γ-selection rules (33) the transition can not be electric dipole but has to be electric quadrupole ($\Delta J = 2$) or a mixture of electric quadrupole and magnetic dipole radiation ($\Delta J = 1$). With this assumption it is then possible to calculate the 0.137 MeV transition intensity from the theoretical conversion data (34) and the experimental data on the intensity of the internal conversion line resulting from this transition. The result found was that 3 % of the disintegrations in In^{116} are followed by a 0.137 MeV transition.

Table 3

The Beta-components of the In^{116} disintegration.

Component	Energy in MeV	Intensity	Half-life in seconds	ft	Group[30]
1	1.00 ± 0.01	51 %	6.37×10^3	16.7×10^4	OB
2	0.87 ± 0.01	28 %	1.15×10^4	18.2×10^4	OB
3	0.60 ± 0.01	21 %	1.58×10^4	6.8×10^4	OB

This result could, as regards order of magnitude, be checked by comparing the intensities of the 0.406 MeV and 2.090 MeV photolines with that of the 1.085 MeV photo-line. This showed agreement within the limits of error.

A further check of the disintegration scheme was provided by comparing the intensities of the 1.274 and 1.085 MeV photo-lines. This was in agreement with the ratio 75/54 given by the term scheme within the limits of error. An accurate comparison, however, was unfortunately not possible because the 1.085 MeV line lies on a very steep Compton slope.

Assuming the disintegration scheme of Fig. 3, it is possible to determine the internal conversion coefficients of the γ-rays of 0.406, 1.085 and 1.274 MeV. The experimental results, together with the theoretically predicted values (35) are given in Table 4. According to these results the experimental conversion coefficients for all three lines agree best with the theoretical values if we consider them all to be electric quadrupole. As the great majority of conversion

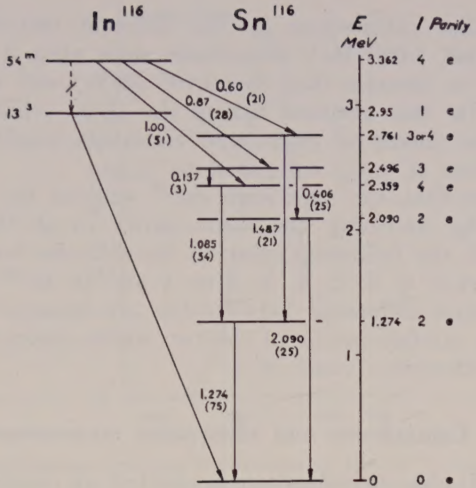

Fig. 3. Proposed disintegration scheme for In^{116}.

Table 4

Internal conversion data.

γ-energy MeV	Electric Multipole		Magnetic Multipole		Experimental Values
	$l = 1$	$l = 2$	$l = 1$	$l = 2$	
0.406	3.49×10^{-3}	1.17×10^{-2}	1.23×10^{-2}	4.34×10^{-2}	1.0×10^{-2}
1.085	4.20×10^{-4}	9.62×10^{-4}	13.15×10^{-4}	29.2×10^{-4}	8.4×10^{-4}
1.274	3.06×10^{-4}	6.79×10^{-4}	8.40×10^{-4}	18.9×10^{-4}	5.7×10^{-4}

lines are electric quadrupole, this may be considered to be additional evidence for the correctness of the term scheme proposed.

The total energy of the disintegration of In^{116} is thus 3.36 ± 0.05 MeV. Absorption measurements (17) gave 2.95 ± 0.10 MeV for the maximum energy of the β-particles emitted in the disintegration of the 13 sec. isomeric state. One has to conclude therefore, that 54 min. isomeric state is due to an excited level in In^{116} situated 410 ± 150 KeV above the 13 sec. ground level. It has not been possible to detect any direct transition between the two levels. That the 0.137 MeV conversion line in the β-spectrum is not due to any isomeric transition is proved by the big K to L ratio of this line. The fact that it was possible to observe rather prominent K and L photo-lines corresponding to a γ-ray of this energy, further proves that the conversion coefficient for this radiation is too small for it to be due to an isomeric transition. Similar arguments rule out even more definitely the possibility of the 0.406 MeV γ-radiation being due to an isomeric transition.

A calculation (35) of the life time of the excited isomeric level in In^{116}, using possible values of (410 + 150) and (410 — 150) KeV for the transition energy, shows that an »effective» l-value of at least 4 must be assumed for this transition. Similar calculations for the different transition probabilities for the 0.137, 0.406 and 2.090 MeV transitions show that if the term scheme is accepted, one has to assume that the 0.406 MeV γ-ray is emitted *before* the 2.090 MeV γ-ray. In this respect, taking the three γ-lines in question to be quadrupole lines, the theory of Segrè and Helmholz would give 0.4 % for the ratio of the intensities of $\gamma_{0.137}$ compared to $\gamma_{2.09}$.

If we now assume that the even-even Sn^{116} nucleus has zero spin, we get a consistent scheme by ascribing the same parity to all the different levels in In^{116} and Sn^{116}, and the following spins to the different levels in Sn^{116}, starting from the ground level: 0, 2, 2, 4, 3, 3 or 4 and in In^{116} 1, 4. Even though the angular momentum difference between the two isomeric levels in In^{116} would thus be only 3, the similar parity of the two states makes this transition parity forbidden with an effective l-value of 4.

6. Coincidence and absorption measurements

In the course of this investigation a number of coincidence measurements were made. These supplied valuable information both for drawing up the decay scheme and for testing its validity.

In the beginning, after the energies of the principal γ-rays had been determined, a series of measurements were made on the $\gamma\gamma$-coincidence effect, using calibrated gold tubes in a standard set-up. These experiments showed a $\gamma\gamma$-coincidence effect $C_{\gamma\gamma}/N_{\gamma} = (1.05 \pm 0.1) \times 10^{-3}$. In the same set-up Co^{60}, which has two γ-rays in cascade with energies of roughly the same value as the main In^{116} γ-rays, showed a $\gamma\gamma$-coincidence effect of $C_{\gamma\gamma}/N_{\gamma} = 0.76 \times 10^{-3}$. It seemed probable therefore that In^{116} should have roughly the equivalent of two γ-rays in cascade per disintegration, or a little more than two in cascade. This is actually the case.

The $\beta\gamma$-coincidence effect showed satisfactory agreement with the disintegration scheme. The corrected ratio $C_{\beta\gamma}/N_{\beta} = (1.86 \pm 0.1) \times 10^{-3}$ found experimentally is to be compared to the ratio $C_{\beta\gamma}/N_{\beta} = 1.80 \times 10^{-3}$, which was computed on the basis of the disintegration scheme and the known effectivity of the γ-tube for the different γ-rays involved.

The position of the 0.137 MeV transition in the scheme could be localized by taking $e^{-}\beta$-coincidences (36) in the intermediate image spectrometer between focused internal conversion electrons of the 0.137 MeV transition, and the continuous spectrum as measured by a β-tube situated directly behind the source. Different absorbers could be introduced between the β-tube and the source.

Fig. 4 shows the »coincidence» line, i.e. the number of $e^{-}\beta$ coincidences per β-particle as a function of the momentum of the electrons focused by the spectrometer.

In Fig. 5 the number of $e^{-}\beta$-coincidences per β-particle as a function of the absorber thickness, is shown. The coincidence rate is equal to the difference between the coincidence counting rate on top of the line, and that next to the line. The last named rate would correspond to chance coincidences $\beta\gamma$-coincidences etc. Although it was not possible to obtain very good statistics because of the short half life of In^{116}, the general appearance of the curve is exactly what one would expect from the decay scheme. In the beginning the rate of $C_{e^{-}\beta}/N_{\beta}$ increases with absorber thickness because the absorption of the 0.60 MeV

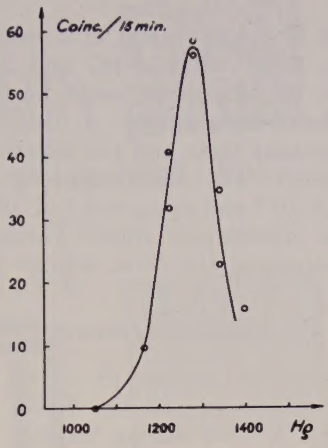

Fig. 4. Number of coincidences between β-particles of the In^{116} continuum and 0.137 MeV transition conversion electrons as a function of the momentum of the focused electrons.

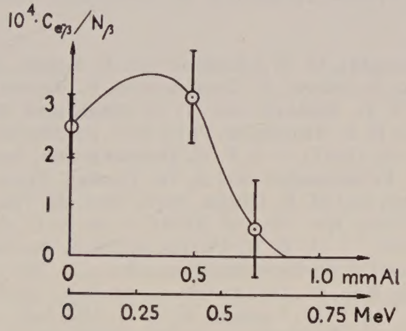

Fig. 5. Number of beta-conversion electron coincidences per β-particle as a function of the absorber thickness between the source and β-tube. The energy scale gives the maximum energy of the β-particles absorbed by the absorber.

component not in coincidence with the 137 keV line decreases N_β more rapidly than the absorption of the 0.87 MeV component decreases $C_{e^-\beta}$. When the 0.60 MeV component has been completely absorbed, further absorption of the 0.87 MeV component affects the coincidence rate directly. When this component had namely almost completely been absorbed at 1.11 mm Al, corresponding to a β-ray cut-off at 0.80 MeV no coincidences were observed, although the β-tube was still counting a relatively large number of β-particles belonging to the 1.00 MeV component. This proves conclusively that the 0.137 MeV γ-transition is not in cascade with either the 0.60 MeV or the 1.00 MeV β-components.

SUMMARY

The disintegration of 54 min. In^{116} has been investigated. It decays with the emission of three β-components with maximum energy 1.00, 0.87 and 0.60 MeV

with relative intensities of 51 %, 28 % and 21 % resp. The resulting Sn^{116} emits six γ-rays of 0.137, 0.406, 1.085, 1.274, 1.487 and 2.090 MeV. No γ-ray at 1.8 MeV, as reported by some investigations, could be found. If existent it would follow less than 3 % of the disintegrations. A disintegration scheme is proposed which satisfies the experimental facts and the selection rules for β- and γ-decay. Internal conversion coefficients were determined for 3 of the γ-rays viz. $\alpha_{0.406} = 1.0 \times 10^{-2}$, $\alpha_{1.085} = 8.4 \times 10^{-4}$ and $\alpha_{1.274} = 5.7 \times 10^{-4}$. These figures agree with that expected for electric quadrupole lines. Various coincidence experiments were performed and substantiated the term scheme proposed.

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